Atomic Clocks and Variations of the Fine Structure Constant

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We describe a new test for possible variations of the fine structure constant α by comparisons of rates between clocks based on hyperfine transitions in alkali atoms with different atomic number Z. H-maser, Cs, and Hg^+ clocks have a different dependence on α via relativistic contributions of order $(Z\alpha)^2$. Recent H-maser vs Hg^+ clock comparison data improve laboratory limits on a time variation by 100-fold to give $\dot{\alpha}/\alpha \leq 3.7 \times 10^{-14}$/yr. Future laser cooled clocks (Be^+, Rb, Cs, Hg^+, etc.), when compared, will yield the most sensitive of all tests for $\dot{\alpha}/\alpha$.

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Since Dirac's large number hypothesis (LNH) [1], the search for a time variation of the fundamental constants has been the subject of much work [2]. Dirac noticed that the ratio of the electrostatic to gravitational forces between an electron and proton ($\sim 2 \times 10^{39}$) was close to the age of the Universe expressed in units of the light transit time across the classical electron radius, $R_e/c = e^2/m_e c^3$. He conjectured that these two very large quantities were proportional, hence, the ratio $e^2/Gm_p m_e$ would vary with the age of the Universe. A fractional change $\delta G/G \approx -5 \times 10^{-11}$/yr would result assuming a universe 2×10^{10} yr old. Teller and co-workers [2,3] have postulated a relationship for the fine structure constant $\alpha^{-1} \sim \ln(hc/Gm_e^2)$, where $(hc/Gm_e^2)^{1/2} \sim$ (electron Compton wavelength)/(Planck length). Taken with the Dirac hypothesis of a time varying G, α may vary $\delta\alpha/\alpha \sim \alpha(\delta G/G) \sim 3.6 \times 10^{-13}$/yr.

Variation of the nongravitational constants is forbidden in general relativity and other metric theories of gravity, where gravitational fields are described as a geometrical property of space-time. The equivalence principle forms the basis for all metric theories and requires local position invariance: In local freely falling frames the outcome of any local nongravitational test experiment is independent of where and when in the Universe it is performed [4]. A changing fine structure constant α, as predicted in some cosmological string theories [5], would violate the equivalence principle signaling the breakdown of gravitation as a geometrical phenomenon and, as we show in this paper, would lead to a drift in the relative frequencies of H masers, Rb, Cs, Hg^+, etc. clocks.

Several analyses of paleontological, geophysical, and astronomical data were made apparently ruling out the LNH variation [2] though there have been conflicting claims for a measured variation of the gravitational constant [6]. The paleontological arguments were based upon the realization that even a small departure of the gravitational constant G from the present day value would make the Earth inhospitable to life. Arguments of this sort have arisen largely as a response to Dirac's LNH and have led to the development of the anthropic cosmological principle (ACP). Accordingly [7], the large number ratio (LNR) values are not a consequence of the above proportionality postulated by Dirac but rather, the present day LNR values are one of a relatively small subset (of all possible LNR values) which will lead to the development of observers, i.e., physicists, astronomers, etc.

The experimental search for a temporal variation of α is divided into what might be called cosmological and modern measurements. For example, a stringent limit on α variation follows from an analysis of isotope ratios ^{149}Sm/^{147}Sm in the natural uranium fission reaction that took place some 2×10^9 yr ago at the present day site of the Oklo mine in Gabon, West Africa [2,8]. This ratio is 0.02 rather than 0.9 as in natural samarium from the neutron flux onto ^{149}Sm during the uranium fission. It is thus deduced that the neutron capture cross section in ^{149}Sm has not changed significantly in 2×10^9 yr from its present day value. Recent modeling [8] of this process has relaxed the original stringent limits by 100-fold to $\dot{\alpha}/\alpha \leq 10^{-15}$. This limits the integrated change in α over the cosmological time period of 2×10^9 yr. In a similar way, astronomical measurements of multiple spectral lines (with different dependence on α and other atomic constants) from a common source with a large cosmological redshift, have been used to place limits on variations of α over cosmological time periods of $\dot{\alpha}/\alpha \leq 4 \times 10^{-12}$/yr [9].

Modern or laboratory measurements are based on clock comparisons with ultrastable oscillators of different physical makeup such as the superconducting cavity oscillator vs cesium hyperfine clock transition [10] or the Mg fine structure transition vs the cesium hyperfine clock transition [11]. Unlike the results inferred from phenomena taking place over cosmological time scales, the clock comparisons are repeatable and are of the duration months to years. These measurements rely on the ultrahigh stability of the atomic standards and set limits a few orders of magnitude less stringent than the cosmological measurements [2,8]. The modern clock comparisons are really complementary

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to the cosmological determinations because they place a limit on a present day variation of α [12].

The string theory prediction [5] for a temporal variation of the fundamental constants has provided a renewed incentive for improved tests of the constancy of α. This paper describes a new method for determining limits on the variation of α by comparing rates for clocks based on atoms of different atomic number Z. The method is based on the increasing importance of relativistic contributions to the hyperfine energy splitting as atomic number Z increases in the group I alkali elements and alkalilike ions. The contribution is a function of αZ which grows faster than $(Z\alpha)^2$ for the heavier atoms and thus differs for hydrogen ($Z = 1$), beryllium ion ($Z = 4$), rubidium ($Z = 37$), cesium ($Z = 55$), and mercury ion ($Z = 80$). Any variation in α, whether a cosmological time variation or a spatial variation via a dependence of α on the gravitational potential [13], will force a variation in the relative clock rates between any pair of these clocks.

We begin by comparing the theoretical expressions for the hyperfine splitting (hfs) in hydrogen and the alkali atoms and ions. All continuously operated microwave atomic frequency standards (H, Rb, Cs, and Hg^+) are based on transitions between ground state hyperfine levels determined by the interaction of a nuclear magnetic moment with the magnetic moment of an $S_{1/2}$ state valence electron. The hydrogen hfs is the simplest and to lowest order in α and m_e/m_p, the splitting used as the clock transition in the H maser is $a_s = \frac{8}{3}\alpha^2 g_p (m_e/m_p) R_\infty c$, where g_p is the proton g factor, m_e and m_p are the electron and proton masses, and $R_\infty c$ is the Rydberg constant in frequency units.

The theory of the hyperfine splitting in alkali atoms and ions is not so well developed as that for hydrogen but much work has been done and the theoretical expressions predict the splittings for the Cs and Hg^+ clock transition frequencies to the 1% level [14]. The full expression for the hyperfine interaction constant A_s [14,15] is

$$A_s = \frac{8}{3}\alpha^2 g_I Z \frac{z^2}{n_*^3}\left(1 - \frac{d\Delta_n}{dn}\right) F_{\mathrm{rel}}(\alpha Z)\,(1-\delta) \times (1-\epsilon)\,\frac{m_e}{m_p}\,R_\infty c\,.$$

The transition frequency between the $I \pm \frac{1}{2}$ states is $(I + \frac{1}{2})A_s$, where I is the nuclear spin angular momentum quantum number.

This expression is composed of several factors. The value of the valence electron wave function at the nucleus, obtained by solving the nonrelativistic Schrödinger equation, is given by the semiempirical Fermi-Segrè formula [16] $\Psi_n^2(0) = (Zz^2/\pi a_0^3 n_*^3)[1 - d\Delta_n/dn]$, where Z is the atomic number, z is the net charge of the remaining ion following the removal of the valence electron, and n_* is the effective quantum number chosen to match the measured energy levels E_{n^*}, for the alkali atom according to the Rydberg formula $E_{n^*} = -z^2\mathrm{Ry}/n_*^2$. $\Delta_n = n - n_*$ is the quantum defect for the nth state. The term $1 - \delta$ is the correction for the departure of the atomic potential from pure Coulomb as the electron enters the relatively large high Z nucleus with $\delta \approx 4\%$ for Cs and 12% for Hg [14]. $1 - \epsilon$ is a similar correction for the finite size of the nuclear magnetic dipole moment with $\epsilon \approx 0.5\%$ for Cs and 3% for Hg [14].

The Casimir correction factor $F_{\mathrm{rel}}(\alpha Z)$ [14,15,17] is obtained when the relativistic wave equation is solved to evaluate the electron wave function in the vicinity of the nucleus. For an $S_{1/2}$ state electron $F_{\mathrm{rel}}(\alpha Z) = 3[\lambda(4\lambda^2 - 1)]^{-1}$, where $\lambda = [1 - (\alpha Z)^2]^{1/2}$ showing F_{rel} is a strong function of α for high Z nuclei. For $\alpha Z \ll 1$, $F_{\mathrm{rel}} \approx 1 + 11(\alpha Z)^2/6$ but with heavier atoms this approximation breaks down since for Cs, $F_{\mathrm{rel}} = 1.39$ and for Hg, $F_{\mathrm{rel}} = 2.26$.

A time variation in α will therefore induce a change in the frequency of an H maser relative to the frequency of a heavy atom hfs transition according to

$$\frac{d}{dt}\ln\left(\frac{A_{\mathrm{alkali}}}{a_{\mathrm{hydrogen}}}\right) = \alpha\frac{d}{d\alpha}\ln(F_{\mathrm{rel}})\left(\frac{1}{\alpha}\frac{d\alpha}{dt}\right).$$

We have assumed the integers z and Z remain constant. Supposing that α changes, there will be a corresponding change in the effective quantum number n_* since it is determined by the Rydberg levels of the valence electron. However, because $n_*^2 \sim E_n/z^2\mathrm{Ry} \sim [1 + \text{higher order in } (z\alpha)^2]$ its changes are small. The finite nuclear volume correction δ does contain terms of order $(\alpha Z)^2$ but its overall sensitivity to α is $\leq 10\%$ of that of F_{rel} and is negligible.

The above ratio between hyperfine transitions in different atoms contains no electron to proton mass ratio and the nuclear g factors enter as a ratio unlike the clock comparisons described in Refs. [10,11]. The above equation is rewritten as

$$\alpha\frac{d}{d\alpha}\ln(F_{\mathrm{rel}}) = (\alpha Z)^2\,\frac{12\lambda^2 - 1}{\lambda^2(4\lambda^2 - 1)} \equiv L_d F_{\mathrm{rel}}(\alpha Z)\,.$$

The sensitivity to α variations $L_d F_{\mathrm{rel}}(\alpha Z)$ is plotted against atomic number Z in Fig. 1.

By analogy with a Dirac particle, the ratio g_I/g_p (g values of a bound nucleon to a free nucleon) is relatively insensitive to α. The nuclear g factors are defined as a ratio of the measured nuclear magnetic moment to the nuclear magneton $eh/2m_p c$ and are determined primarily by the strength of the strong interaction. For an electron bound to a nucleus of charge Z there is a relativistic mass contribution to the electron g factor of order $(\alpha Z)^2$ [15]. By contrast, the strong force binding a nucleon in a nucleus "saturates," i.e., remains relatively constant with increasing atomic number unlike the electromagnetic binding of an electron to a nucleus. We therefore assume there is no corresponding contribution to the nuclear g-factor ratio which grows with atomic number Z as strong as the $(\alpha Z)^2$ dependence of F_{rel}.

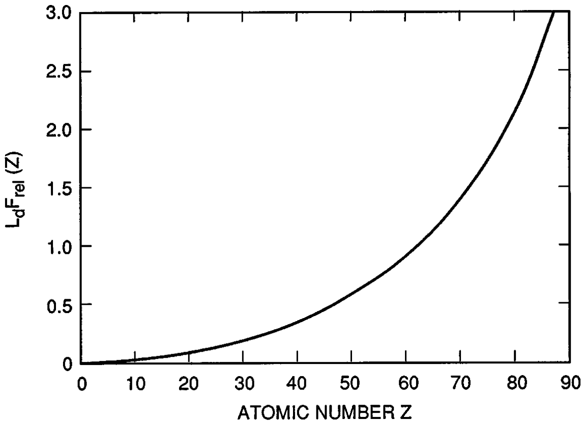

FIG. 1. The function $L_d F_{\mathrm{rel}}(Z)$ plotted against atomic number Z.

As above, for the comparison of two clocks, each based on a transition in different alkali atoms with $Z > 1$, there will be a relative drift in rates

$$\frac{d}{dt} \ln \frac{A_{\mathrm{alkali1}}}{A_{\mathrm{alkali2}}} = [L_d F_{\mathrm{rel}}(Z_1) - L_d F_{\mathrm{rel}}(Z_2)] \frac{1}{\alpha} \frac{d\alpha}{dt}.$$

Table I shows the size of the sensitivity $L_d F_{\mathrm{rel}}(Z_1) - L_d F_{\mathrm{rel}}(Z_2)$ for various clock intercomparisons that might be used to detect a temporal variation in α (or spatial with d/dt replaced by d/dU, where U is the solar gravitational potential [13]). A larger sensitivity would cause a larger clock rate difference given a nonzero value for $\dot{\alpha}/\alpha$. Alternatively, given a variation in α, the six distinct drift rates of Table I would predict a clear signature which would be useful in discriminating against systematic errors that might show up in any single intercomparison. For example, the Cs vs Hg^+ rate difference should be $1.4/0.74 \approx 1.9$ times greater than the H maser vs Cs rate difference, etc.

Several clock comparisons have been made which can be used to search for a variation of α. Long term comparisons of Cs to H-maser clocks are carried out in the generation and maintenance of the worldwide atomic time scale (TAI). A recent comparison carried out over a 1 yr period between two cavity autotuned active H masers and the primary cesium standards, CS1 and CS2 (at PTB in Braunschweig, Germany) showed a 1.5×10^{-16}/day relative frequency drift [18]. Similar clock comparisons have been made at the U.S. Naval Observatory [19] with comparable clock rate drifts. Since $L_d F_{\mathrm{rel}}(55) = 0.74$ we find $\dot{\alpha}/\alpha \leq 1.5 \times 10^{-16}$/day–$0.74 = 7 \times 10^{-14}$/yr.

We have developed [20,21] an ultrastable frequency standard based on Hg^+ ions confined to a linear ion trap, and have recently completed a 140 day clock rate comparison (to be published) between it and a cavity tuned H maser [22]. In that comparison, a limit of $2.1(0.8) \times 10^{-16}$/day was established for the frequency drift between these two long term stable clocks. The Allan deviation of this clock comparison is shown in Fig. 2. This is a more sensitive test for α variations than the Cs vs H maser comparison since $L_d F_{\mathrm{rel}}(80) = 2.2$ and establishes an upper bound $\dot{\alpha}/\alpha \leq 3.7 \times 10^{-14}$/yr.

This Hg^+ vs H maser limit represents a tenfold improvement over the recent limit [11] and rules out the LNH variation of α ($\sim 3.6 \times 10^{-13}$/yr) discussed in the introduction. It should be noted that these results are the *only present day laboratory* tests with enough sensitivity to rule out such variations. The limits established in Ref. [11] on an α variation ($\leq 2.7 \times 10^{-13}$/yr) were inferred from astrophysical limits placed on $\alpha^2 g_p m_e/m_p$ [9] over a time interval of almost 10^{10} yr.

The Hg^+ vs H maser results presented here represent a 100-fold improvement over the best laboratory limits ($\leq 4 \times 10^{-12}$/yr) established in the superconducting cavity vs Cs frequency comparisons of Ref. [10]. This improvement follows from the very good long term stability of the atomic Hg^+ and H-maser clocks, with relative drift $\sim 10^{-16}$/day, as compared to the superconducting cavity oscillator where instrumental drifts can lead to frequency drifts of a few parts in 10^{14}/day [10].

In summary, we have developed a new method for detecting variations of the fine structure constant α by examining relative drift rates between atomic clocks

TABLE I. The sensitivity of various clock rate comparisons to a variable fine structure constant. The entry is $L_d F_{\mathrm{rel}}(Z_1) - L_d F_{\mathrm{rel}}(Z_2)$ and converts fractional changes in α to a drift in clock rates between the two given clocks. For example, if $\dot{\alpha}/\alpha = 10^{-14}$/yr, a frequency drift of 2.2×10^{-14}/yr between an H maser and an Hg^+ clock would result.

	H	Rb	Cs	Hg^+
H	0	0.3	0.74	2.2
Rb	−0.3	0	0.45	1.9
Cs	−0.74	−0.45	0	1.4
Hg^+	−2.2	−1.9	−1.4	0

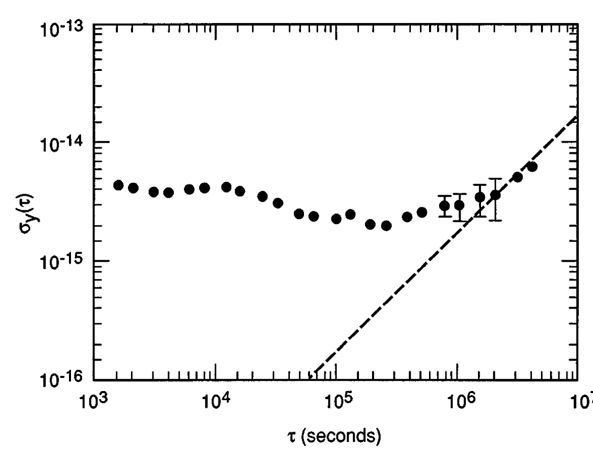

FIG. 2. The measured Allan deviation for the 140 day H maser vs Hg^+ clock comparison. The dashed line at 45° is the linear drift estimate $2(\pm 1) \times 10^{-16}$/day.

which are continuously monitored in time scales in several laboratories worldwide. We have searched for such drifts in a clock comparison between Hg^+ and H-maser clocks and improved modern day limits on an α variation by 2 orders of magnitude. Further improvements will follow as laser cooled Be^+, Rb, Cs, and Hg^+ [23] microwave standards are developed. Comparisons of their clock rates should establish the most sensitive search for any temporal variation of α and may reach a sensitivity approaching the string theory predictions [5]. Finally, this method also shows that comparisons between Cs, Hg^+, Rb, Be^+, and H-maser clocks can be used to improve the complementary search for a dependence of α on the gravitational potential [13].

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[1] P. A. M. Dirac, Nature (London) **139**, 323 (1937).

[2] F. J. Dyson, *Aspects of Quantum Theory* (Cambridge University Press, Cambridge, 1972), pp. 213–236; *Current Trends in the Theory of Fields* (AIP, New York, 1983), pp. 163–168; B. W. Petley, *The Fundamental Physical Constants and the Frontier of Measurements* (Adam Hilger, Boston, 1985).

[3] E. Teller, Phys. Rev. **73**, 801 (1948); L. D. Landau, in *Niels Bohr and the Development of Physics,* edited by W. Pauli (Pergamon Press, London, 1955); B. S. DeWitt, Phys. Rev. Lett. **13**, 114 (1964); C. J. Isham, A. Salam, and J. Strathdee, Phys. Rev. D **3**, 1805 (1971).

[4] C. M. Will, *Theory and Experiment in Gravitational Physics* (Cambridge University, Cambridge, 1981).

[5] T. Damour and A. M. Polyakov, Nucl. Phys. **B423**, 532–558 (1994).

[6] T. C. Van Flandern, *Precision Measurements and Fundamental Constants II,* NBS Circular No. 617 (U.S. GPO, Washington, DC, 1981), p. 625; R. D. Reasonberg, Philos. Trans. R. Soc. London A **310**, 227 (1983).

[7] J. D. Barrow and F. J. Tipler, *The Anthropic Cosmological Principle* (Oxford University Press, New York, 1988).

[8] A. I. Schlyakter, Nature (London) **25**, 340 (1976); P. Sisterna and H. Vucetich, Phys. Rev. D **41**, 1034 (1990).

[9] J. N. Bachall and M. Schmidt, Phys. Rev. Lett. **19**, 1294 (1967); A. M. Wolfe, R. L. Brown, and M. S. Roberts, Phys. Rev. Lett. **37**, 179 (1976).

[10] J. P. Turneaure and S. Stein, *Atomic Masses and Fundamental Constants V* (Plenum, London, 1976), pp. 636–642.

[11] A. Godone, C. Novero, P. Travella, and K. Rahimullah, Phys. Rev. Lett. **71**, 2364 (1993).

[12] B. W. Petley, in *Quantum Metrology and Fundamental Physical Constants,* edited by P. H. Cutler and A. A. Lucas, NATO ASI Ser. B, Vol. 98 (Plenum, New York, 1983), pp. 333–351.

[13] J. P. Turneaure, C. M. Will, B. F. Farrel, E. M. Mattison, and R. F. C. Vessot, Phys. Rev. D **27**, 1705 (1983).

[14] H. Kopfermann, *Nuclear Moments* (Academic Press, New York, 1958), pp. 123–138; H. B. G. Casimir, *On the Interaction between Atomic Nuclei and Electrons* (Freeman, San Francisco, 1963), p. 54.

[15] M. Mizushima, *Quantum Mechanics of Atomic Spectra and Atomic Structure* (W. A. Benjamin, New York, 1970).

[16] L. L. Foldy, Phys. Rev. **111**, 1093 (1958).

[17] C. Schwartz, Phys. Rev. **97**, 380 (1955).

[18] N. A. Demidov, E. M. Ezhov, B. A. Sakharov, B. A. Uljanov, A. Bauch, and B. Fisher, in *Proceedings of the 6th European Frequency and Time Forum, Noordwijk, The Netherlands, 1992* (European Space Agency, Noordwijk, 1992), pp. 409–414.

[19] L. A. Breakiron, in *Proceedings of the 25th Annual Precise Time Interval Applications and Planning Meeting,* NASA Conference Publication No. 3267 [U.S. Naval Observatory Time Service Department (TSS1), Washington, DC, 1993], pp. 401–412.

[20] J. D. Prestage, G. J. Dick, and L. Maleki, J. Appl. Phys. **66**, 1013 (1989); J. D. Prestage, R. L. Tjoelker, G. J. Dick, and L. Maleki, J. Mod. Opt. **39**, 221–232 (1992).

[21] R. L. Tjoelker, J. D. Prestage, G. J. Dick, and L. Maleki, in *Proceedings of the 1994 IEEE Frequency Control Symposium* (IEEE, New York, 1994), pp. 739–743.

[22] Sigma Tau Standards, Tuscaloosa, AL 35403.

[23] M. E. Poitzsch, J. C. Bergquist, W. M. Itano, and D. J. Wineland, in *Proceedings of the 1994 IEEE Frequency Control Symposium* (Ref. [21]), p. 744; A. Clairon *et al.* (to be published).